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«Жанармай, катализ және электрохимия институты» АҚ

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ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ НАУК
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NAS RK is pleased to announce that News of NAS RK. Series of chemistry and technologies scientific journal has been accepted for indexing in the Emerging Sources Citation Index, a new edition of Web of Science. Content in this index is under consideration by Clarivate Analytics to be accepted in the Science Citation Index Expanded, the Social Sciences Citation Index, and the Arts & Humanities Citation Index. The quality and depth of content Web of Science offers to researchers, authors, publishers, and institutions sets it apart from other research databases. The inclusion of News of NAS RK. Series of chemistry and technologies in the Emerging Sources Citation Index demonstrates our dedication to providing the most relevant and influential content of chemical sciences to our community.

Қазақстан Республикасы Үлттық гылым академиясы «ҚР ¥ҒА Хабарлары. Химия және технология сериясы» ғылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бүл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Webof Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын үсынады. ҚР ¥ҒА Хабарлары. Химия және технология сериясы Emerging Sources Citation Index-ке енуі біздің қогамдастық үшін ең өзекті және беделді химиялық ғылымдар бойынша контентке адалдығымызды білдіреді.

НАН РК сообщает, что научный журнал «Известия НАН РК. Серия химии и технологий» был принят для индексирования в Emerging Sources Citation Index, обновленной версии Web of Science Содержание в этом индексировании находится в стадии рассмотрения компанией Clarivate Analytics для дальнейшего принятия журнала в the Science Citation Index Expanded, the Socia1 Sciences Citation Index и the Arts & Humanities Citation Index. Web of Science предлагает качество в глубину контента для исследователей, авторов, издателей и учреждений. Включение Известия НАН РК в Emerging Sources Citation Index демонстрирует нашу приверженность к наиболее актуальному и влиятельному контенту по химическим наукам для нашего сообщества.

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DEVELOPMENT OF THE CONDITIONS FOR STORING THALLIUM AMALGAM

Abstract. It has been established that lNH_2SO_4 solution and distilled water can be used as a protective medium for a long-term storage (30-40 days) of thallium amalgam of eutectic composition (8.6 mas.%), without sliming of its surface and an insignificant change of its concentration. An inverse dependence between the decomposition reaction rate and the ratio of the solution volume per unit of the contact surface (V:S), which corresponds to the height of the liquid layer (h) above the amalgam, is observed. At h ~ 2.5 cm and higher, the reaction rate becomes stabilized. The value of metal loss during its storage for 10-15 days does not exceed 0.5-1.0 mas.% of the initial thallium content in the amalgam.

It has been shown that the rate of dissolution of thallium from amalgam is rather low, and in sulfuric acid solution it is only slightly higher than that in water with all S:V ratios. A similar picture is observed upon thallium amalgam decomposition in the same media in an open vessel; the difference consists only in a higher K value (Table 4). Thus, with S:V = 1:2.5, in an open vessel K in lNH_2SO_4 makes up 10,5.1011 mol/cm2.sec, and in H_2O – 7.3.1011 mol/cm2.sec, whereas in a closed vessel K in lNH_2SO_4 makes up 1,9.1011 mol/cm$_2$.sec, and in H_2O – 1.5.1011 mol/cm2.sec.

For 40 days of thallium amalgam (8.54 mas.%) storage in 1NH_2SO_4 the loss of thallium has been 1.3 mas.%, and in the distilled water - 1 mas.%. The quantity of the dissolved metal increases in proportion to the experiment duration, which is characteristic for a zero-order reaction, and this dependence is observed for all S:V ratios from 1:1 to 1:7.

Besides, a possibility to use some saturated hydrocarbons, wherein no sliming of thallium amalgam occurs, as a protective wash liquid has been shown.

Key words: thallium amalgam, chemical preparation, storage, decomposition, sulfuric acid, water, wash liquid.

Introduction. Thallium is a very important metal in both theoretical and practical terms, combining the properties of both alkali and heavy (Pb, Ag, Au) metals. It is highly soluble in mercury, which is very important for amalgam hydrometallurgy (at 293 K - 43 mas.%), and for solving other technical problems.

The development of structures and technologies for manufacturing mercury-thallium thermometers, hermetic contacts, contactrons and thermal contactrons is complicated by the fact that there are no reliable data on thallium amalgam permanency in water solutions, methods of controlling the content of metal in amalgam and methods of processing the amalgam wastes, formed in the process of filling the stocks.

The literature data on thallium amalgam permanency in various media are rather limited. The basic data, related mainly to the behavior of amalgam in the water solutions, have been obtained upon the development of a technology for thallium production by the methods of amalgam hydrometallurgy [1-6]. It has been found that in some cases amalgam sliming occurs. This is facilitated by the presence in the solution of the substances - emulsifiers, which are not sulfur-containing compounds or metal oxides, formed due to oxidation, for example, by air oxygen. Small droplets of amalgam are enveloped with these substances or particles, which form stable adsorption salts on the surface, which contributes to the fragmentation of thallium amalgam [2].

The work practice with thallium amalgam shows that sliming occurs when amalgam is in the air, due to metal oxidation by air oxygen with the formation of oxides on the surface. The behavior of thallium amalgam in acidic aqueous solutions has been studied in the works [7-9]. It has been established that the decomposition rate does not depend on the concentration in amalgam and its oxidation proceeds under the effect of dissolved oxygen in the studied acidic solutions, which by the speed of an effect upon thallium amalgam are arranged in the following series:

$$CH_3COOH<CH_2ClCOOH<H_2O_2<HCl<HBr<HI$$

It is seen from the presented series that in inorganic acids thallium is oxidized to a greater extent than in organic ones. Though, even upon the effect of inorganic acids on amalgam the process of its decomposition proceeds at low rates. Thus, for example, the reaction of amalgam decomposition in 0.01 n H_2SO_4 proceeds at a not-too-high rate (K-1,6.10^{-3} mol/cm^2.sec.). The authors [9] have also found that the dissolution of thallium proceeds also rather slowly upon the treatment of amalgam in 1H_2SO_4 and distilled water.

It can be assumed on this basis, that thallium amalgam of high concentrations has a rather high permanency in acids due to the low rate of metal dissolution. However, this conclusion is of a general character, and for specific purposes additional studies for the development of optimal conditions for storing amalgam are required.

Materials and methods. The amalgam permanency test was carried out by holding it for different periods of time at the room temperature under a layer of water and acidic solution, both in the open and closed vessels. When conducting the experiment the solution samples were periodically (at set time intervals) taken for the quantitative determination of thallium ions therein. Based on these data, the concentration of thallium, remaining in the amalgam, and the degree of its depletion at a given time were calculated. Herewith, in the course of the experiment, the ratio of the solution volume to the quantity of the taken amalgam, and such an indicator as Sam:Vsol- the quantity of the solution per unit of the mercury alloy surface were measured.

The calculations of the kinetic parameters were carried out by the known methods.

The permanency of thallium amalgam in 1NH_2SO_4 and distilled water was studied in most detail. In contrast to the previous studies [9], it was found that thallium amalgam decomposition in 1NH_2SO_4 proceeded at a low rate. The metal loss value during its storage period for 10–15 days did not exceed 0.5–1.0 mas.% of the initial thallium content in amalgam.

An increase in the storage time to 19, 29 days resulted in a more significant decrease in thallium concentration (3.5–5 mas.%). After 40 days of storage of thallium amalgam (8.54 mas.%) in 1NH_2SO_4 the loss of thallium was 1.3 mas.%, and in distilled water - 1 mas.%.

Results and discussion. The results are shown in Table 1, from which it is clear that thallium amalgam decomposition in 1n sulfuric acid proceeds at a slow rate. The metal loss value during the storage period for 10-15 days does not exceed 0.5-1.0 mas.% of the initial thallium content in amalgam. An increase in the storage time to 19, 29 days results in a more significant decrease in thallium concentration (3.5–5 mas.%).

Table 1 – Thallium amalgam permanency upon holding in 1NH_2SO4 in a closed vessel

Holding time, day	Sam: Vsolution	CTI, mas.%	Degree of TI loss, %	Note
0	1:1.70	8.36	00	Sam = 41 T = 297K
5	1:1.20	8.32	0.50	
9	1:0.61	8.22	1.15	
15	1:0.22	8.19	2.22	
19	1:0.10	8.09	3.50	
29	1:0.07	7.98	4.97	
0	1:2.50	9.26	00	Sam = 41 T = 293K
5	1:2.40	9.24	0.13	
10	1:1.82	9.21	0.30	
15	1:1.60	9.20	0.55	
23	1:1.37	9.17	0.97	
28	1:1.00	9.14	1.30	
35	1:0.84	9.10	1.72	
42	1:0.81	9.08	2.10	
48	1:0.79	9.01	2.84	
65	1:0.76	8.90	4.09	
0	1: 5.00	8.44	0	S = 10,2 T = 293K
5	1:3.50	8.42	0.29	
10	1:2.50	8.40	0.59	
14	1:1.00	8.37	0.86	
17	1:0.50	8.28	2.02	

It has turned out that with Sam:Vsolution=1:1 the oxidizer can be consumed in less than a day, with Sam:Vsolution= 1:2.5; 1:5.0; 1:7 - for 3.8 and 15 days, respectively. Since the experiments for amalgam decomposition have been carried out for a longer time (30–40 days), it can be concluded that the consumed oxygen is continuously replenished through its absorption from the air.

Table 2 shows the experimental data for determining thallium amalgam permanency in distilled water and, for comparison, in lNH_2SO_4

under the same conditions. As can be seen, in the studied solutions a rather high amalgam permanency is observed. For 40 days of storage of thallium amalgam (8.54 mas.%) in lNH_2SO_4 the loss of thallium has been 1.3 mas.%, and that in distilled water - 1 mas..%.

Table 2 – Thallium amalgam permanency upon holding in $1NH_2SO_4$ and distilled water in a closed vessel with Sam:Vsol= 1:2.5 (t=301K; Sam=10.2 cm^2)

Holding time, day	$1NH_2SO_4$		Distilled H_2O	
	CTI, mas.%	TI, mas.%	CTI, mas.%	Tl,- мас.%
0	8.640	0	8.640	0
8	8.622	0.23	8.623	0.15
15	8.612	0.35	8.617	0.32
20	8.592	0.61	8.604	0.46
25	8.582	0.74	8.594	0.58
29	8.572	0.86	8.587	0.67
35	8.560	1.01	8.575	0.83
40	8.539	1.28	8.553	1.01

As mentioned above, an inverse dependence between the decomposition reaction rate and the ratio of the solution volume per unit of the contact surface (V:S), which corresponds to the height of the liquid layer (h) above the amalgam, is observed. At h ~ 2.5 cm and higher, the reaction rate becomes stabilized.

Fig. 1 shows the change in the quantity of thallium ions in the studied liquids over time upon amalgam decomposition (8.64 mas.% Tl).

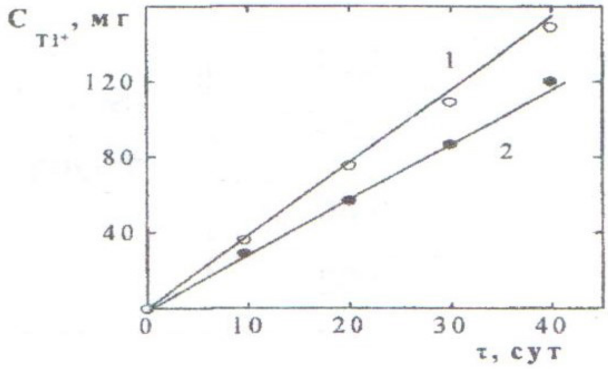

Fig.1. Dependence of the content of thallium ions in the solution on the time period of amalgam (8,54 mas.%) decomposition in $1NH_2SO_4$ (1) and H_2O (2) Sam: Vsol=1:2.5; Vsol= 25cm^3

As is seen, the quantity of the dissolved metal increases in proportion to the experiment duration, which is characteristic of a zero-order reaction, and this dependence is observed with all S:V ratios from 1:1 to 1:7.

Table 3 presents the values of the constants of decomposition rates (K), calculated by us for thallium amalgams in the closed vessels.

Table 3 – Constants of decomposition rates of thallium amalgam in $1H_2SO_4$ and H_2O in a closed vessel with different S:V ratios (t=301KSam=10.17 cm^2; Ctl = 8.64 mas.%)

K.1011, mol/cm^2.sec		Sam: Vsol
1 NH_2SO_4	H20	
3.56±0.25	3.35±0.05	1:1.0
1.89±0.11	1.48±0.09	1:2.5
1.57±0.14	1.40±0.17	1:4.0
154±0.08	1.22±0.10	1:5.0
1.11±0.09	1.04±0.14	1:7.0

The rate of dissolution of thallium from amalgam, as it follows from the Table, is rather low, and in sulfuric acid solution it is only slightly higher than that in water with all S:V ratios.

Besides, it has been found that the value of the solution volume above the amalgam surface has a significant effect on the process rate: with an increase of V by S = 1 cm^2, the value of K decreases. A similar picture is observed upon thallium amalgam decomposition in the same media in an open vessel; the difference consists only in a higher K value (Table 4). Thus, with S:V = 1:2.5, in an open vessel K in lNH_2SO_4 makes up 10,5.1011 mol/cm^2.sec, and in H_2O – 7.3.1011 mol/cm2.sec, whereas in a closed vessel K in lNH_2SO_4 makes up 1,9.1011 mol/cm^2.sec, and in H_2O – 1.5.1011 mol/cm^2.sec.

Table 4 – Permanency of thallium amalgams upon holding it in lNH_2SO_4 and distilled water in an open vessel with Vam=const; Sam=10.17 cm^{2}t=297K

Holding time, day	H_2SO_4 S:V=1:2.5		1 NH_2SO_4			
	CTI, mas. %	Degree of loss, mas.%	S:V=1:2.5		S:V=1:4	
			CTI, mas.%	Degree of loss, mas.%	CTI, mas.%	Degree of loss, mas.%
0	7.24		0.16		7.26	
1	7.23	0.16	0.29	0.23	7.25	0.05
2	7.22	0.29	0.52	0.40	7.24	0.27
5	7.20	0.52	0.77	0.81	7.23	0.34
6	7.19	0.77	1.76	1.12	7.22	0.56
12	7.12	1.76	3.75	2.35	7.18	1.11
21	6.99	3.75	3.69	4.56	7.06	3.06
56	6.00	3.69	0.16	5.08	6.97	4.37

Conclusion. The presented data prove that thallium amalgam decomposition proceeds due to the interaction of metal with oxygen, dissolved in the water solutions. Based on the reference data on oxygen solubility, cm3/cm3:0.283 in l H_2SO_4 and 0.252 in H_2O at 298 K, the quantity of gas, dissolved in the volumes of the used solutions, has been calculated, and the time during which the initial content of oxygen in the solution may be completely consumed, has been determined, accounting for the reaction rate. It has turned out that with S:V = 1:1 the oxidizer can be consumed in less than a day, and with S:V = 1: 2.5:1:5.0; 1:7 - for 3, 8 and 15 days, respectively. Since the experiments for amalgam decomposition have been carried out for a longer time (30–40 days), it can be concluded that the consumed oxygen is continuously replenished through its absorption from the air.

Besides, a possibility to use some saturated hydrocarbons, wherein no sliming of thallium amalgam occurs, as a protective wash liquid has been studied. With this purpose the amalgam permanency in pentane, chloroform, methylene chloride has been studied. It has been established that while holding thallium amalgam 7.72 mas.% in these liquids for 10 minutes, its decomposition practically does not take place. The amalgam, washed and dried with chloroform and methylene chloride, has been placed in a glass ampoule, which then has been evacuated, filled with argon and sealed. The observations of the state of amalgam in the ampoule have shown that after washing with chloroform the amalgam surface remains clean, without a film, and a dark rim appears on the glass after 24 hours, at the height of the amalgam level, which does not change with time.

After washing the amalgam with methylene chloride, after 2 hours, a turbidity appears on the amalgam surface and glass, which spreads over the entire inner surface of the ampoule. Upon the further amalgam storage (for a month or more), the picture does not change: a rather clean surface of the amalgam in the ampoules and after washing it with distilled water, followed by the removal of moisture in a vacuum.

The liquids, tested by us for washing the amalgam, apart from methylene chloride, provide quite satisfactory surface cleanliness, for the preservation of which later, upon placing the amalgam in the glass vessels, it is necessary, first and foremost, to exclude entering of oxygen into the system (equipment, ampoules, etc.).

Thus, it follows from the results of the study that lNH_2SO_4 solution and distilled water, with a height of the liquid layer of 2.5 cm and higher, can be used as a protective medium for a long-term storage (30-40 days) of thallium amalgam of eutectic composition, without sliming of its surface and an insignificant change of its concentration.

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ТАЛЛИЙ АМАЛГАМАНЫ САҚТАУ ШАРТТАРЫН ЖАСАУ

Аннотация. lNH_2SO_4 және тазартылған судың ерітіндісін эвтектикалық құрамы бар таллий амальгамын ұзақ сақтаған кезде (30-40 тәулік) қорғаныс ортасы ретінде қолдануға болатындығы анықталды (8,6 масс.%). Оның бетін тегістемей және мәнін өзгертпейтін концентрация. Ыдырау реакциясының жылдамдығы мен ерітіндінің көлем бірлігінің жанасу бетіне қатынасы (V:S) арасында кері байланыс байқалады, бұл сұйықтық қабатының амальгамадан жоғары биіктігіне (h) сәйкес келеді. H ~ 2,5 см және одан жоғары болғанда реакция жылдамдығы тұрақталады. Сақтау кезінде металдың ысырап мөлшері 10-15 тәулік ішінде амальгамадағы таллийдің бастапқы құрамынан% 0,5-1,0 аспайды.

Таллийдің амальгамнан еру жылдамдығы едәуір төмен екендігі және күкірт қышқылы ерітіндісінде барлық S:V қатынаста суға қарағанда сәл ғана жоғары екендігі көрсетілген. Таллий амальгамасының сол ортада ашық ыдыста ыдырауы кезінде ұқсас сурет байқалады, айырмашылық тек К-нің жоғары мәнінде болады (4-кесте). Сонымен, S:V = 1:2,5 ашық ыдыста, lNH_2SO_4-те K 10,5.1011 моль / $см^2$ • с, ал H_2O-да 7,3 • 1011 моль / $см^2$ • с, ал lNH_2SO_4 ішіндегі тұйық ыдыста K болады 1,9.1011 моль / $см^2$ с, ал H_2O-да - 10,5.1011 моль / $см^2$ с.

Таллий амальгамасын сақтаудың 40 күнінде (8,54 масс.%) lNH_2SO_4-де таллийдің жоғалуы 1,3 мас.%, ал тазартылған суда - 1%. Ерітілген металдың мөлшері эксперименттің ұзақтығына пропорционалды түрде көбейеді, бұл нөлдік тәртіптегі реакцияға тән және бұл тәуелділік 1:1-ден 1:7-ге дейінгі барлық S:V қатынастарында байқалады.

Сонымен қатар, таллий амальгамасының қабығы жоқ кейбір қаныққан көмірсутектерді пайдалану үшін жууға арналған сұйықтық ретінде қолдану мүмкіндігі көрсетілген.

Түйін сөздер: таллий амальгамасы, химиялық препарат, сақтау, ыдырау, күкірт қышқылы, су, жуу сұйықтығы.

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РАЗРАБОТКА УСЛОВИЙ ХРАНЕНИЯ ТАЛИЕВОЙ АМАЛЬГАМЫ

Аннотация. Установлено, что раствор lNH_2SO_4 и дистиллированная вода могут быть использованы в качестве защитной среды при длительном хранении (30-40 суток) амальгамы таллия эвтектического состава (8,6 мас.%), без шлифования ее поверхности и незначительного изменение его концентрации. Наблюдается обратная зависимость между скоростью реакции разложения и соотношением объема раствора на единицу контактной поверхности (V:S), которое соответствует высоте жидкого слоя (h) над амальгамой. При h ~ 2,5 см и выше скорость реакции стабилизируется. Величина потерь металла при его хранении в течение 10-15 суток не превышает 0,5-1,0 мас.% от исходного содержания таллия в амальгаме.

Показано, что скорость растворения таллия из амальгамы довольно мала и в растворе серной кислоты лишь немного выше, чем в воде при всех соотношениях S:V. Аналогичная картина наблюдается при разложении амальгамы таллия в тех же средах в открытом сосуде, различие состоит только в более высоком значении K (табл. 4). Так, при S: V = 1: 2,5 в открытом сосуде K в lNH_2SO_4 составляет 10,5.1011 моль / см^2 · с, а в H_2O - 7,3 · 1011 моль / см^2 · с, тогда как в закрытом сосуде K в lNH_2SO_4 составляет 1,9.1011 моль / см^2 сек, а в H_2O - 10,5.1011 моль / см^2 сек.

За 40 суток хранения амальгамы таллия (8,54 мас.%) В lNH_2SO_4 потеря таллия составила 1,3 мас.%, а в дистиллированной воде - 1 мас.%. Количество растворенного металла увеличивается пропорционально продолжительности эксперимента, что характерно для реакции нулевого порядка, и эта зависимость наблюдается для всех соотношений S: V от 1: 1 до 1: 7.

Кроме того, показана возможность использования некоторых предельных углеводородов, при которых не происходит шелушения амальгамы таллия в качестве защитной промывной жидкости.

Ключевые слова: амальгама таллия, химическое приготовление, хранение, разложение, серная кислота, вода, промывная жидкость.

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PHYTONCIDES IN THE COMPOSITION OF COMMON BIRD CHERRY

Abstract. Everyone knows that forest air is very good for health, and one of the most important reasons for this is the presence of phytoncides in it, which kill or suppress pathogens and have a healing effect. Also, phytoncides are one of the factors of the natural immunity of plants (plants sterilize themselves with the products of their vital activity). Their large number is allocated by plants. One of them is the common bird cherry. Cherry-a representative of the genus of plums of the Rosaceae family. The view includes low trees and shrubs. Cheremukha-forest orderly. Its flowers and leaves are rich in phytoncides, thanks to which they exude an alluring aroma. However, when they break down, they release prussic acid, which is dangerous for all living things. This gave them the opportunity to attract and destroy pests. Phytoncides are volatile biologically active substances formed by plants that kill or inhibit the growth and development of bacteria, microscopic fungi, and protozoa. In addition to all of the above, bird cherry has exceptional properties. The strong, somewhat intoxicating scent of flowers and leaves cleanses the air of germs. Antimicrobial properties of phytoncides have led to a large number of studies on their use in medicine, veterinary medicine, plant protection, storage of fruit and vegetable products, in the food industry and other areas of practice.

Almost all parts of the plant have bactericidal, fungicidal and insecticidal properties. In folk medicine, bird cherry has long been used as an astringent, fixing, anti-inflammatory and anti-scurvy agent. Bird cherry produces the most powerful phytoncides containing prussic acid. Protozoa die under the influence of bird cherry phytoncides in 5 minutes. On the basis of numerous studies, the time of death of protozoa after non-contact exposure to phytoncidal plants has been established. Especially a lot of phytoncides are released by young leaves in spring and summer, in autumn phytoncides are released much less.

The presence of tannins and essential oil in the fruit has an anti-inflammatory effect, which is used to treat inflammatory processes in the gastrointestinal tract and dysentery. The infusion of cherry fruits has a destructive effect on microorganisms. Preparations of the fruits of the common cherry have an antiseptic effect. They are used in dental practice in the treatment of inflammatory processes of the oral mucosa, paradontosis, toothache and hypovitaminosis.

Key words: Bird cherry, phytoncides, plant, bacteria, protozoa, useful properties.

Introduction.

Bird cherry is a representative of the Plum genus of the Rosaceae family. The species includes low trees and shrubs. The trunk of the plant is covered with dark gray bark with occasional rusty and brown spots. The young leaves are green, glistening with gold. The old ones are dull. The flowers on the pedicels are small, white and very fragrant. The fruit is a black drupe. Bird cherry is a forest orderly. Its flowers and leaves are rich in phytoncides, thanks to which they exude an alluring aroma. However, when they split, they secrete prussic acid, which is dangerous for all living things. This gave them the ability to attract and kill pests.

Cherry Berries contain a complex of antioxidants, which makes them indispensable for those who seek

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